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(54) **Adsorbents for hydrogen recovery by pressure swing adsorption**

(57) Hydrogen is purified from a feed gas mixture including hydrogen and at least one impurity selected from carbon monoxide and nitrogen using an adsorption apparatus having a discharge end adsorption layer containing an adsorbent with a Henry's law constant ( $K_H$ ) at 70°F (21°C) for the impurity from 0.8 to 2.2 mmole/g/atm (0.1 MPa) for carbon monoxide or from 0.55 to 1.4 mmole/g/atm (0.1 MPa) for nitrogen. High purity (99.99+%) hydrogen can be obtained.

**EP 1 097 746 A2**

## Description

**[0001]** This invention relates to adsorption processes, and more particularly to hydrogen production via pressure swing adsorption (PSA) and vacuum swing adsorption processes.

**[0002]** Hydrogen production via pressure swing adsorption ( $H_2$  PSA) is a multi-million dollar industry supplying high purity hydrogen for chemical producing industries, metals refining and other related industries. Typical commercial sources for the production of hydrogen are the reforming of natural gas or partial oxidation of various hydrocarbons. Other hydrogen-rich gas sources which can be upgraded by PSA technology to a high purity product include refinery off-gases with  $C_1$ - $C_{10}$  hydrocarbon contaminants. See, e.g., US-A-3,176,444 to Kiyonaga. The reforming is carried out by reacting the hydrocarbon with steam and/or with oxygen-containing gas (e.g., air or oxygen-enriched air), producing a hydrogen gas stream containing accompanying amounts of oxides of carbon, water, residual methane and nitrogen. Unless it is desired to recover carbon monoxide, the carbon monoxide is customarily converted to carbon dioxide by water gas shift reaction to maximize the hydrogen content in the stream. Typically, this gas stream is then sent to a PSA system.

**[0003]** In a typical PSA system, a multicomponent gas is passed to at least one of multiple adsorption beds at an elevated pressure to adsorb at least one strongly sorbed component while at least one component passes through. In the case of  $H_2$  PSA,  $H_2$  is the most weakly adsorbed component which passes through the bed. At a defined time, the feed step is discontinued and the adsorption bed is depressurized in one or more concurrent steps which permits essentially pure  $H_2$  product to exit the bed with a high recovery of the most weakly adsorbed component,  $H_2$ . Then a countercurrent desorption step is carried out, followed by countercurrent purge and repressurization.

**[0004]** The cost of hydrogen from integrated reformer/PSA systems is impacted by both the capital and operating costs of the system. Clearly, economic production of hydrogen requires minimization of operating and capital costs. Capital cost is most widely affected by the size of the reformer and the size of the PSA beds. PSA bed size decreases as the feed loading (lb-moles (kg-moles) of feed gas processed/bed volume) of the PSA increases. Feed loading can be increased by either improved process cycles or improved adsorbents. The size of the reformer is impacted mostly by the hydrogen recovery of the PSA. Improvements in hydrogen recovery in the PSA result in smaller reformer size (the reformer does not need to produce as much hydrogen because of better recovery in the PSA). Improvements in hydrogen recovery also lead to a reduced demand for reformer feed gas, i.e., natural gas, which constitutes the largest operating cost of the reformer. Hydrogen recovery in the PSA can also be improved by either improved process cycles or improved adsorbents.

**[0005]**  $H_2$  PSA process performance (on-line time, feed loading, product purity, recovery) is usually dictated by the second most weakly adsorbing component in the  $H_2$ -rich stream. A bed can stay on feed, producing pure  $H_2$ , only until the level of impurity breakthrough reaches the desired product purity. For steam/methane reformer (SMR) cases, the PSA feed gas composition is typically about 1%  $N_2$ , 5%  $CH_4$ , 5%  $CO$ , 18%  $CO_2$  and the remainder  $H_2$ . To produce high purity  $H_2$  (99.99+%) with this feed gas composition,  $N_2$  is the key breakthrough component since it is the most weakly adsorbing feed gas component besides  $H_2$ . Since  $N_2$  is the key breakthrough component, it has been common to place a zeolite adsorbent with high capacity for  $N_2$  at the product end of the bed. In some cases, the  $H_2$  purity spec is 99.9% with less than 10 ppm  $CO$  in the product  $H_2$ . In these cases, the plant becomes  $CO$ -controlling and zeolites are the prior art adsorbents for  $CO$  removal from  $H_2$ .

**[0006]** For example, US-A-3,430,418 to Wagner teaches a layered adsorption zone with the inlet material comprising activated carbon and the discharge end containing zeolite for the removing the minor component of  $N_2$ ,  $CO$  or  $CH_4$ . US-A-3,564,816 to Batta exemplifies the use of CaA (5A) zeolite as an adsorbent for PSA processing. US-A-3,986,849 to Fuderer et al. discloses a layered bed adsorption zone with activated carbon at the feed end of the bed and CaA zeolite at the discharge end.

**[0007]** The art teaches a variety of means for removing  $CO$  and/or  $N_2$  from gas mixtures. In particular, Li containing X and Ca containing A type zeolites have been widely employed as adsorbents for separating  $N_2$  or  $CO$  from more weakly adsorbing gas mixtures. See, e.g., US-A-4,813,980, US-A-4,859,217, US-A-5,152,813, US-A-5,174,979, US-A-5,354,360 and US-A-5,441,557, US-A-5,912,422, EP-A-0 855 209 and WO-A-97/45363.

**[0008]** Despite the foregoing developments and their asserted advantages, there is still room for improvement in the art.

**[0009]** Thus, it is desired to provide an improved method for recovering purified hydrogen in  $CO$  and/or  $N_2$  controlled  $H_2$  PSA. It is also desired to provide improved adsorbents and systems for use in the improved method.

**[0010]** It is further desired to provide an improved  $CO$  coldbox offgas purification method. It is also desired to provide improved adsorbents and systems for use in the improved method.

**[0011]** The invention provides an adsorption process to purify hydrogen from a feed gas mixture including hydrogen and at least one impurity selected from carbon monoxide and nitrogen, said process comprising:

providing an adsorption apparatus comprising a discharge end adsorption layer comprising an adsorbent having a

$K_H$  at 70°F (21°C) for said impurity from 0.85 to 1.40 mmole/g/atm (0.1 MPa);  
feeding through said adsorption apparatus said feed gas mixture; and  
collecting a product gas from said adsorption apparatus, wherein said product gas consists essentially of hydrogen.

5 **[0012]** The invention further provides an adsorption process to purify hydrogen from a feed gas mixture including hydrogen and nitrogen, said process comprising:

selecting at least one adsorbent based on said at least one adsorbent having a  $K_H$  at 70°F (21°C) for nitrogen of 0.55 to 1.40 mmole/g/atm (0.1 MPa);  
10 providing an adsorption apparatus comprising a discharge end adsorption layer comprising said at least one adsorbent;  
feeding through said adsorption apparatus said feed gas mixture; and  
collecting a product gas from said adsorption apparatus, wherein said product gas consists essentially of hydrogen.

15 **[0013]** Still further provided is an adsorption process to purify hydrogen from a feed gas mixture including hydrogen and carbon monoxide, said process comprising:

providing an adsorption apparatus comprising a discharge end adsorption layer comprising an adsorbent having a  $K_H$  at 70°F (21°C) for carbon monoxide from 0.8 to 2.2 mmole/g/atm (0.1 MPa);  
20 feeding through said adsorption apparatus said feed gas mixture; and  
collecting a product gas from said adsorption apparatus, wherein said product gas consists essentially of hydrogen.

**[0014]** Apparatuses to perform the process of the invention are also provided.

25 **[0015]** The inventors have discovered that conventional separation methods employing conventional adsorbents in conventional devices can be significantly improved by providing adsorbents having Henry's law constants ( $K_H$ ) within predetermined ranges. The Henry's law constant for an adsorption isotherm is defined as the initial isotherm slope. See, for example, "Physical Adsorption of Gases," D. M. Young and A. D. Crowell, p. 104, (Butterworths, London 1962). The units of the constant are in amount of gas adsorbed per unit weight of adsorbent per unit of pressure (e.g., mole of gas adsorbed/gram of adsorbent/atmosphere (0.1 MPa) of pressure).

30 **[0016]** The invention provides an improved PSA process for the purification of  $H_2$  containing gas streams, wherein the minor impurity which dictates  $H_2$  purity comprises at least one of  $N_2$  and CO. At least the final adsorbent layer for the process comprises an adsorbent having a  $K_H$  at 70°F (21°C) for the minor impurity from 0.85 to 1.40 millimole of impurity/gram of adsorbent/atmosphere (0.1 MPa) of impurity pressure (or simply mmole/g/atm (0.1 MPa)).

**[0017]** In  $N_2$  controlling embodiments, at least the final adsorbent layer comprises a material having a Henry's law constant at 70°F (21°C) for  $N_2$  between 0.55 and 1.40 mmole/g/atm (0.1 MPa), preferably from 0.55 to 0.83 mmole/g/atm (0.1 MPa) and from 0.85 to 1.40 mmole/g/atm (0.1 MPa). In embodiments,  $K_H N_2$  is at least 0.90 mmole/g/atm (0.1 MPa) or at least 1.00 mmole/g/atm (0.1 MPa).

**[0018]** In CO controlling embodiments, at least the final adsorbent layer comprises a material having a Henry's law constant at 70°F (21°C) for CO between 0.8 and 2.2 millimole of CO/gram of adsorbent/atmosphere (0.1 MPa) of CO pressure, preferably from 1 to 2 mmole/g/atm (0.1 MPa), even more preferably, at least 1.5 mmole/g/atm (0.1 MPa).

**[0019]** The feed temperature is preferably 32 to 140°F (0 to 60 °C). The feed pressure is preferably from 100 to 1000 psig (0.7 to 7 MPa). Employing an adsorbent according to the invention under these conditions maximizes the feed loading and recovery of the PSA process over other adsorbents tested. The bulk density is preferably from 30 to 60 lbs/ft<sup>3</sup> (475 to 975 kg/m<sup>3</sup>). The particle diameter is preferably from 0.5 to 3 mm.

45 **[0020]** The final  $H_2$  purity is preferably at least 99.9%, more preferably at least 99.99%, even more preferably at least 99.999%, and most preferably at least 99.9999%.

**[0021]** The apparatus of the invention preferably comprises an inlet end adsorption layer comprising activated carbon, activated alumina, silica gel or combinations thereof, in addition to the discharge end adsorption layer comprising an adsorbent of the invention. It is preferred that the discharge end adsorption layer consist essentially of the adsorbent of the invention. In embodiments, all adsorption layers of the adsorption apparatus consist essentially of an adsorbent of the invention.

**[0022]** For CO controlling processes, the adsorbent is preferably selected from NaX (or 13X) both with and without binder, NaA (or 4A) both with and without binder and potassium exchanged chabazite both with and without binder.

55 **[0023]** For  $N_2$  controlling processes, the adsorbent is preferably selected from CaA (5A) exchanged to greater than 80% calcium levels both with and without binder, sodium exchanged chabazite both with and without binder.

**[0024]**  $H_2$  recovery in accordance with the invention is higher than that of prior art processes wherein the discharge end adsorption layer is substantially devoid of adsorbents of the invention. Preferably,  $H_2$  recovery is at least 75% for both  $N_2$  controlling processes and CO controlling processes.

**[0025]** The invention is suitable for N<sub>2</sub> controlling processes wherein the feed gas comprises hydrogen and 0.1 to 20% nitrogen, and for CO controlling processes wherein the feed gas comprises hydrogen and 0.1 to 40% carbon monoxide.

**[0026]** It will be appreciated by those skilled in the art that the invention additionally facilitates the removal of CO and/or N<sub>2</sub> from gas streams other than H<sub>2</sub> gas streams, such as, e.g., He gas streams, and the removal of impurities from gas streams by vacuum swing adsorption processes as well as pressure swing adsorption processes.

**[0027]** The invention will be illustrated in more detail with reference to the following Examples, but it should be understood that the present invention is not deemed to be limited thereto.

#### EXAMPLE 1

**[0028]** A process development unit (PDU) was used to measure H<sub>2</sub> PSA performance for a feed gas composition comprising: 10.5% CO<sub>2</sub>, 0.2 % N<sub>2</sub>, 5.6 % CH<sub>4</sub>, 3.9 % CO and 79.8 % H<sub>2</sub>. The feed pressure was 400 psig (2.76 MPa) and temperature 70°F (21°C). The beds were filled with 50% activated carbon and 50% zeolite. Using a 5 bed PSA cycle with 3 equalizations, different zeolite adsorbents were screened for process performance. The results of the experiments for the adsorbents tested for a H<sub>2</sub> product with 11 ppm CO were as follows:

Adsorbent	K <sub>H</sub> CO @ 70°F (21°C) (mmole/g/atm (0.1 MPa))	Relative H <sub>2</sub> Recovery (%)	Relative Feed Loading
Standard 5A	2.412	-	1.00
Binderless 13X	2.036	+0.5	1.09

**[0029]** This shows that the H<sub>2</sub> recovery decreases with increasing K<sub>H</sub> CO. The binderless 13X gives both a higher recovery and higher feed loading than Standard 5A (Union Carbide Data Sheet F-2184B, "Linde Molecular Sieve Type 5A"). It is not obvious that standard 5A, with a higher equilibrium capacity for CO at a given pressure, would not perform as well as the lower CO capacity binderless 13X in a H<sub>2</sub> PSA process. In fact, the prior art teaches away from the instant invention in extolling the advantages of using adsorbents that happen to have a higher K<sub>H</sub> CO for H<sub>2</sub> purification, such as LiLSX (K<sub>H</sub> CO of 23.37 mmole/g/atm (0.1 MPa) @ 70°F (21°C)) (WO 97/45363).

#### EXAMPLE 2

**[0030]** An adsorption process simulator was used to estimate H<sub>2</sub> PSA performance for a feed gas composition of 10.5 % CO<sub>2</sub>, 0.2 % N<sub>2</sub>, 5.6 % CH<sub>4</sub>, 3.9 % CO and 79.8 % H<sub>2</sub>. The feed pressure was 400 psig (2.76 MPa) and temperature 70°F (21°C). The beds were filled with 50% activated carbon and 50% zeolite. Using a 5 bed PSA cycle with 3 countercurrent equalizations, different zeolite adsorbents were screened for process performance. All process simulator input parameters other than equilibrium parameters (e.g., density, mass transfer, void fraction) were the same as those used for standard 5A to ensure the results correlate only with differences in equilibrium isotherm parameters between adsorbents. The results of the simulations for adsorbents with a range of K<sub>H</sub> CO for a H<sub>2</sub> product with 11 ppm CO were as follows:

Adsorbent	K <sub>H</sub> CO @ 70°F (21°C) (mmole/g/atm (0.1 MPa))	Relative Feed Loading
LiLSX	23.37	0.94
High Performance 5A Binderless, > 90% Ca	6.339	1.00
Standard 5A	2.412	1.00
Binderless 13X	2.036	1.06
Standard 13X	1.155	1.04
Activated Carbon	0.620	1.01

(continued)

Adsorbent	K <sub>H</sub> CO @ 70°F (21°C) (mmole/g/atm (0.1 MPa))	Relative Feed Loading
HY	0.239	0.84

**[0031]** The results of the simulations for standard 5A and binderless 13X show the same trend as the H<sub>2</sub> PDU results, even though the isotherm parameters were the only adsorbent specific parameters used in the simulation. The prior art adsorbent with the larger K<sub>H</sub> CO, standard 5A, has an inferior relative feed loading to binderless 13X for a CO controlled H<sub>2</sub> PSA process. The process simulation was repeated for adsorbents with a wide range of K<sub>H</sub> CO. Fig. 1 shows a plot of the relative feed loading versus K<sub>H</sub> CO from these simulations. Feed loading increases as the K<sub>H</sub> CO increases to about 2.0 mmole/g/atm (0.1 MPa), creating a non-obvious maximum in performance, after which further increases in the K<sub>H</sub> CO yield lower feed loading. This result is contrary to what would be expected from the prior art, which recommends adsorbents for CO controlled H<sub>2</sub> PSA having K<sub>H</sub> COs in excess of 2.2 mmole/g/atm (0.1 MPa), such as Standard 5A (see, e.g., US-A-3,564,816 and US-A-3,986,849), CaX (see, e.g., US-A-4,477,267) and LiLSX (see, e.g., EP-A-0 855 209 and WO-A-97/45363). The results of these simulations show that there is both a preferred minimum value as well as a preferred maximum value for the K<sub>H</sub> CO where performance begins to degrade.

## EXAMPLE 3

**[0032]** An adsorption process simulator was used to estimate H<sub>2</sub> PSA performance for a feed gas composition of 0.11% CO<sub>2</sub>, 0.10% N<sub>2</sub>, 1.34% CH<sub>4</sub>, 0.5% CO and 97.95% H<sub>2</sub>. The feed pressure was 325 psig (2.24 MPa) and temperature 100°F (38°C). Using a 6 bed PSA cycle, binderless 13X and standard 5A zeolite adsorbents were screened for process performance. The activated carbon and zeolite bed splits were optimized for each zeolite. The actual measured process simulator input parameters (density, mass transfer, void fraction) were used for the adsorbents in this simulation. The results of the simulations for a H<sub>2</sub> product with 1 ppm CO were as follows:

Adsorbent	K <sub>H</sub> CO @ 70°F (21°C) (mmole/g/atm (0.1 MPa))	Relative H <sub>2</sub> Recovery (%)	Relative Feed Loading
Standard 5A	2.412	-	1.00
Binderless 13X	2.036	+0.5	1.09

**[0033]** This Example clearly shows that the advantages of the invention are maintained when the bed splits are optimized for each adsorbent and the effects of differences in process simulator input parameters between adsorbents are included. This Example also shows that the improved performance of binderless 13X over standard 5A extends to a CO cold box effluent type gas stream in addition to a standard SMR stream as shown in Example 1.

## EXAMPLE 4

**[0034]** A process development unit (PDU) was used to measure H<sub>2</sub> performance for a feed gas composition of 14.2% CO<sub>2</sub>, 5.5% N<sub>2</sub>, 4.5% CH<sub>4</sub>, 3.0% CO and 72.8% H<sub>2</sub>. The feed pressure was 446 psig (3.075 MPa) and temperature 70°F (21°C). The beds were filled with 60% activated carbon and 40% zeolite. Using a 5 bed PSA cycle with 3 equalizations, different zeolite adsorbents were screened for process performance. The results of the experiments for adsorbents tested for a H<sub>2</sub> product with 500 ppm N<sub>2</sub> were as follows:

Adsorbent	K <sub>H</sub> N <sub>2</sub> @ 70°F (21°C) (mmole/g/atm (0.1 MPa))	Relative H <sub>2</sub> Recovery (%)	Relative Feed Loading
> 80% CaX	5.256	-0.4	0.94
High Performance 5A Binderless, > 90% Ca	0.844	+0.7	1.08

(continued)

Adsorbent	$K_H N_2$ @ 70°F (21°C) (mmole/g/atm (0.1 MPa))	Relative H <sub>2</sub> Recovery (%)	Relative Feed Loading
Standard 5A	0.429	-	1.00

**[0035]** Fig. 2 is a plot of the PDU relative H<sub>2</sub> recovery versus  $K_H N_2$  for CaX, high performance 5A and standard 5A. A non-obvious maximum in feed loading is observed at a  $K_H N_2$  of 0.8 mmole/g/atm (0.1 MPa). The prior art teaches away from the instant invention in extolling the advantages of using adsorbents for H<sub>2</sub> purification that happen to have lower  $K_H N_2$ , such as standard 5A (see, e.g., US-A-3,564,816 and US-A-3,986,849) or higher  $K_H N_2$ , such as LiLSX (see, e.g., EP-A-0 855 209 and WO-A-97/45363), which has a  $K_H N_2$  of 1.710 mmole/g/atm (0.1 MPa) and CaX (see, e.g., US-A-4,477,267) which has a  $K_H N_2$  of 5.256 mmole/g/atm (0.1 MPa). The PDU tests show that there is an intermediate range of values for the  $K_H N_2$  where performance is unexpectedly enhanced.

#### EXAMPLE 5

**[0036]** An adsorption process simulator was used to estimate H<sub>2</sub> PSA performance for a feed gas composition of 10.5 % CO<sub>2</sub>, 0.2 % N<sub>2</sub>, 5.6 % CH<sub>4</sub>, 3.9 % CO and 79.8 % H<sub>2</sub>. The feed pressure was 400 psig (2.76 MPa) and temperature 70°F (21°C). The beds were filled with 50% activated carbon and 50% zeolite. Using a 5 bed PSA cycle with 3 equalizations, different adsorbents were screened for process performance. All process simulator input parameters (density, mass transfer, void fraction) were the same as those used for standard 5A to ensure the results correlate only with differences in equilibrium isotherm parameters between adsorbents. The results of the simulations for adsorbents with a range of  $K_H N_2$  for a H<sub>2</sub> product with 100 ppm N<sub>2</sub> were as follows:

Adsorbent	$K_H N_2$ @ 70°F (21°C) (mmole/g/atm (0.1 MPa))	Relative H <sub>2</sub> Recovery (%)
Binderless > 80% CaX	6.080	-0.3
LiLSX	1.710	+0.3
High Performance 5A Binderless, > 90% Ca	0.844	+0.6
Standard 5A	0.429	-
Standard 13X	0.306	-0.8

**[0037]** These process simulations, as in the case for CO, allow an optimum range of  $K_H N_2$ , providing superior H<sub>2</sub> PSA performance to be defined. The maximum occurs at a lower  $K_H N_2$  value than both binderless CaX ( $K_H N_2$  of 6.080 mmole/g/atm (0.1 MPa) @ 70°F (21°C)) and LiLSX ( $K_H N_2$  of 1.710 mmole/g/atm (0.1 MPa) @ 70°F (21°C)) which are taught in the prior art. The minimum occurs at a higher  $K_H N_2$  value than standard 5A which is taught in the prior art. Both the simulation and PDU results support the definition of an optimum range of  $K_H N_2$  values which provide superior performance for H<sub>2</sub> PSA processes that is not inclusive of the prior art adsorbents.

**[0038]** The preceding Examples clearly demonstrate that there is an optimum range of adsorbent  $K_H$  for achieving the best performance in both N<sub>2</sub> and CO impurity controlled H<sub>2</sub> adsorption processes.

**[0039]** While the invention has been described in detail and with reference to specific examples thereof, it will be apparent to one skilled in the art that various changes and modifications can be made therein without departing from the scope thereof as defined in the following claims.

#### Claims

1. An adsorption process to purify hydrogen from a feed gas mixture including hydrogen and at least one impurity selected from carbon monoxide and nitrogen, said process comprising feeding said feed gas mixture through an adsorption apparatus comprising a discharge end adsorption layer, characterised in that said discharge end adsorption layer comprises an adsorbent having a  $K_H$  at 21°C (70°F) for carbon monoxide from 0.8 to 2.2 mmole/g/0.1 MPa (atm) or for nitrogen from 0.55 to 0.83 or from 0.85 to 1.40 mmole/g/0.1 MPa (atm).
2. A process of Claim 1, wherein said adsorbent has a  $K_H$  at 21°C (70°F) for said impurity from 0.85 to 1.40

mmole/g/0.1 MPa (atm).

3. A process of Claim 1 or Claim 3, which is carbon monoxide controlled pressure swing adsorption.
- 5 4. A process of Claim 1 or Claim 3, wherein said impurity is carbon monoxide and said adsorbent has a  $K_H$  at 21°C (70° F) for carbon monoxide from 0.8 to 2.2 mmole/g/0.1 MPa (atm).
5. A process of Claim 4, including the step of selecting said adsorbent based on having said  $K_H$  value.
- 10 6. A process of Claim 4 or Claim 5, wherein said adsorbent has a  $K_H$  at 21°C (70°F) for carbon monoxide from 1 to 2 mmole/g/0.1 MPa (atm).
7. A process of Claim 6, wherein said adsorbent has a  $K_H$  at 21°C (70°F) for carbon monoxide from 1.5 to 2 mmole/g/0.1 MPa (atm).
- 15 8. A process of any one of Claims 4 to 7, wherein said feed gas comprises hydrogen and 0.1 to 40% carbon monoxide.
9. A process of any one of Claims 4 to 7, wherein said product gas comprises at least 99.9%  $H_2$ .
- 20 10. A process of any one of Claims 4 to 9, wherein said adsorbent is selected from sodium exchanged X zeolite with and without binder, sodium exchanged A zeolite with and without binder and potassium exchanged chabazite with and without binder.
- 25 11. An adsorption process to purify hydrogen from a feed gas mixture including hydrogen and nitrogen, said process comprising feeding said feed gas mixture through an adsorption apparatus comprising a discharge end adsorption layer, characterised in that said discharge end adsorption layer comprises an adsorbent which has been selected based on having a  $K_H$  at 21°C (70° F) for nitrogen of 0.55 to 1.40 mmole/g/0.1 MPa (atm).
- 30 12. A process of Claim 1, wherein said impurity is nitrogen, or Claim 11, wherein said adsorbent has a  $K_H$  at 21°C (70°F) for nitrogen from 0.55 to 0.83 mmole/g/0.1 MPa (atm) or from 0.85 to 1.40 mmole/g/0.1 MPa (atm).
13. A process of any one of Claims 1, 11 or 12, which is nitrogen controlled pressure swing adsorption.
- 35 14. A process of Claim 13, wherein said adsorbent has a  $K_H$  at 21°C (70°F) for nitrogen of at least 0.9 mmole/g/0.1 MPa (atm).
15. A process of Claim 14, wherein said adsorbent has a  $K_H$  at 21°C (70°F) for nitrogen of at least 1 mmole/g/0.1 MPa (atm).
- 40 16. A process of any one of Claims 11 to 15, wherein said feed gas comprises hydrogen and 0.1 to 20% nitrogen.
17. A process of any one of Claims 11 to 16, wherein said product gas comprises at least 95.0%  $H_2$ .
- 45 18. A process of any one of Claims 11 to 17, wherein said adsorbent is selected from greater than 80% calcium-exchanged A zeolite with binder, and sodium-exchanged chabazite with and without binder.
19. A process of any one of the preceding claims, wherein a feed temperature is 32 to 140°F (0 to 60°C) and a feed pressure is 0.7 to 7 MPa (100 to 1000 psig).
- 50 20. A process of any one of the preceding claims, wherein said adsorbent has a bulk density of 475 to 975 kg/m<sup>3</sup> (30 to 60 lbs/ft<sup>3</sup>).
21. A process of any one of the preceding claims, wherein said adsorbent has a particle diameter of 0.5 to 3 mm.
- 55 22. A process of any one of the preceding claims, wherein said discharge end adsorption layer consists essentially of said adsorbent.

23. A process of any one of the preceding claims, wherein an inlet end adsorption layer of said adsorption apparatus consists essentially of activated carbon, activated alumina, silica gel and combinations thereof.

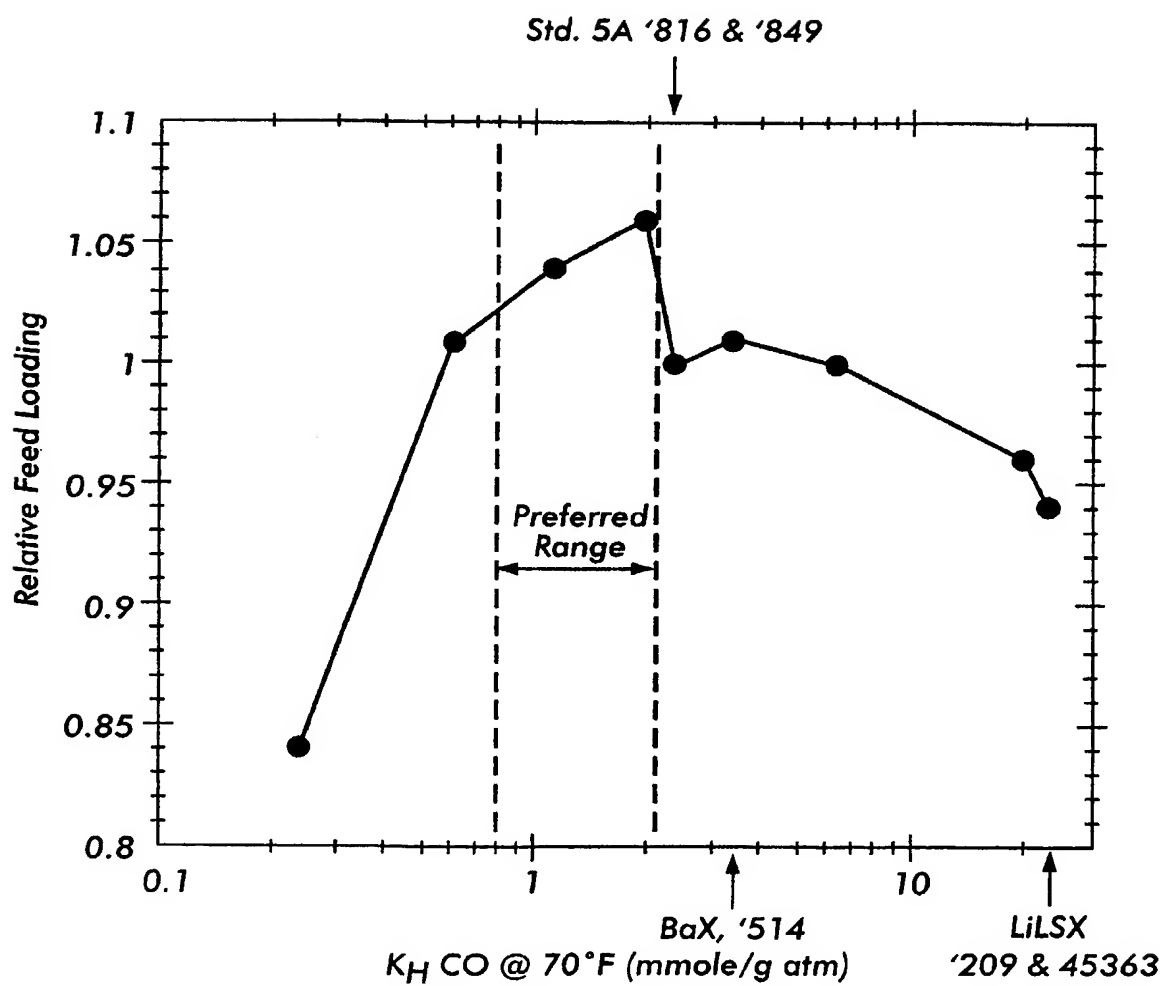
24. A process of any one of the preceding claims, wherein all adsorption layers of said adsorption apparatus consist essentially of said adsorbent.

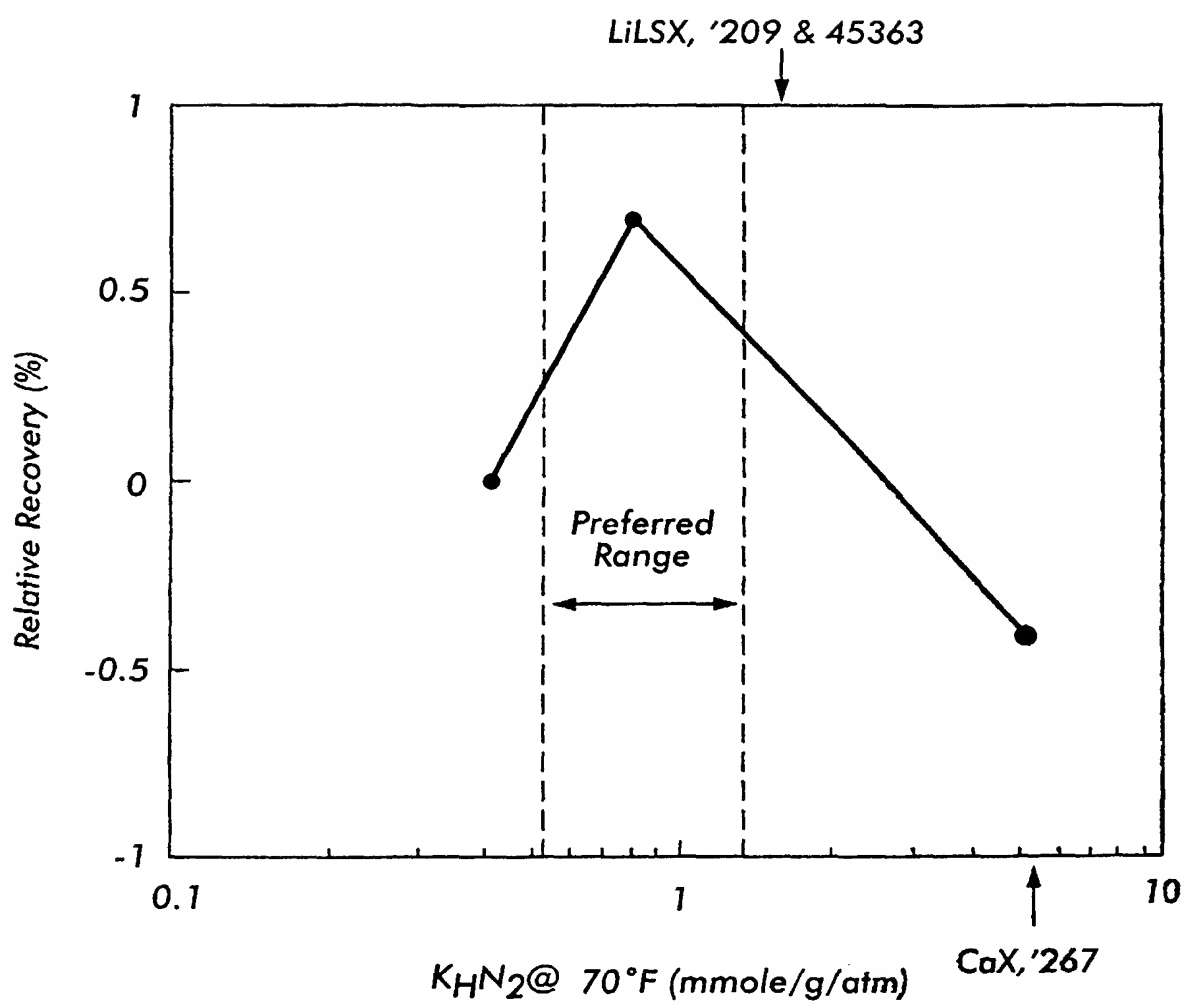
25. The use, to improve hydrogen recovery from a feed gas mixture including hydrogen and at least one impurity selected from carbon monoxide and nitrogen by purification in an adsorption apparatus, of an adsorbent having a  $K_H$  at 21°C (70°F) for carbon monoxide from 0.8 to 2.2 mmole/g/0.1 MPa (atm) or for nitrogen from 0.55 to 0.83 or from 0.85 to 1.40 mmole/g/0.1 MPa (atm) in a discharge end adsorption layer of said apparatus.

26. A use of Claim 20, wherein the adsorbent and/or process parameters are as defined in any one of Claims 2 to 24.

27. An adsorption apparatus for use in a process as defined in Claim 1, said apparatus comprising a discharge end adsorption layer, characterised in that said discharge end adsorption layer comprises an adsorbent having a  $K_H$  at 21°C (70°F) for carbon monoxide from 0.8 to 2.2 mmole/g/0.1 MPa (atm) or for nitrogen from 0.55 to 0.83 or from 0.85 to 1.40 mmole/g/0.1 MPa (atm).

28. An apparatus of Claim 27 adapted for carrying out a process as defined in any one of Claims 2 to 24.

**FIG. 1**

**FIG.2**